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## The Investigation of Parameters of the Stimulated Mossbauer Radiation Emitted under Conditions of Controllable Nuclear Spin Dynamics on the basis of Real Magnetic Materials (Final Report)

### Introduction

Mossbauer spectroscopy presents a unique possibility for studying coherent dynamics of nuclear spins in solids. The theoretical and experimental works on the investigations of quasienergies of the nuclear spin under NMR conditions [1-3] serve as a confirmation. The most successful experiments of such type were performed lately [4,5]. The interest to Mossbauer investigations in a radiofrequency (RF) excitation (nonresonant, in general) regime retains up to the date [6,7]: two applicable points maintain that interest. First, the object under investigation is as a rule magnetically ordered substances wherein hyperfine field on the nucleus is directly related with the behavior of local magnetization, that is an important feature of magnetically ordered materials in RF fields. Second is the possible influence of RF modulated medium upon the properties of propagating wave: numerous alike effects (such as self-induced transparency, inversionless amplification [8] ) are well known in optics. The problems considered in this work are closely connected to such effects in gamma-optics. We are interested in changes in the spectrum and in temporal dependence of probability of gamma-transitions (spontaneous and stimulated) under conditions of controllable dynamics of nuclear spins in magnetic materials. The study contains the following parts: 1. Magnetodynamics in magnetic materials under the conditions of RF modulation, 2. The method of quasienergetic spin states in gamma transition analysis, 3. The discussion of some possibilities and concluding remarks.

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## Magnetodynamics in the external alternating fields.

Experimental studies of RF modulation of Mossbauer spectra in magnetic materials were carried out in a lot of works [3-5, 9-11]. A great deal of them concerns the case of external oscillating fields of high (compared to Larmour frequency of a nuclear spin) frequency. Nevertheless there were also some attempts to observe the spectra with changing in direction (rotation) of the external field at low (compared to Larmour frequency) frequency [9]. This case of low frequency fields is attractive in the sense that according to the theory [7] despite the large-scale variations of hyperfine field the collapse of structure does not occur, as it would be at high frequencies. Thus, in the low frequency mode, one has a possibility of controllable variation of the positions of spectral lines even at fields of high amplitude. Coherent influence on a Mossbauer nucleus by RF field also leads to the temporal modulation of its susceptibility in gamma-range [12]. This quite familiar fact acquires the special sense in the domain of low frequency modulation. In this event, the period of modulation is large enough to get for this while the desired gamma-optical effect.

The problem of RF modulation of gamma-transitions includes first of all the technique of generating the alternating fields with large amplitude on the nucleus. For isotope of iron Fe-57, wherein hyperfine magnetic interaction provides an amplification of external RF field to two, three orders, it is not too hard to get the alternating fields on the nucleus of 10-30 Koe even in magnetically hard substances. Such fields are sufficient for resonant effects to be observed. Speaking about non-resonant effects, in particular, low-frequency modulation of spectrum, observable transformations are possible only in the regime of total magnetization reversal. In this case we are forced to deal with magnetically soft substances. The same problem can be solved using magnetic systems of "easy plane" type, e.g. FeBO<sub>3</sub> (see below).

But the presence of magnetic anisotropy fields (due to crystallography origin or to the sample shape) results in some difficulties of generating the alternating hyperfine fields of desired type. Although a large number of Mossbauer investigations of magnetic materials under RF excitation was carried out, the processes of transformation of external RF field into the field on nuclei taking into consideration the peculiarities of real magnetic systems are not studied in detail. We are familiar with only one work attempting to solve this problem [13].

We have done an analysis of regimes of the magnetization motion in magnetic materials. The task of that analysis could be formulated like this. What is the dependence of dynamical behavior of the magnetization on the frequency and amplitude of alternating field. In other words, whether at all frequencies of the alternating field the magnetization follows the alternating field coherently. This question

becomes important for rotating fields. Stating of such a question is prompted by the adiabatic theorem [14], known in nuclear magnetism, according to which the magnetization follows the alternating field only if angular velocity of its direction changing is much less than the frequency of Larmour precession related to amplitude of the alternating field. In this, adiabatic, case the magnetization will display a precession around the instant direction of the external field. Dissipative processes will force for the magnetization to coincide with the instant field direction. In such motion, the hyperfine field has approximately the same temporal dependence as the external field does. However, the condition of adiabaticity could be broken down either due to increasing the frequency of external field or due to decreasing its amplitude. The same may be the case due to crystal anisotropy fields and internal fields of the sample. In this case the magnetization does not catch up with the external field. Such a regime cannot be used for the controllable modulation of hyperfine field. We analyze the ferromagnetic system using the simple equations of magnetization  $M$  motion of Landau-Lifshitz type:

$$\frac{dM}{dt} = \gamma[MH] - \frac{\alpha}{M^2}[M[MH]], \text{ where}$$

$H = H_e(t) + H_a + H_i$  - the sum of external magnetic field, field of magnetic anisotropy and internal field in sample,  $\gamma$ ,  $\alpha$  - gyromagnetic ratio and dissipation coefficients.

First, we consider isotropic case ( $H_a = 0$ ,  $H_i = 0$ ).  $H_e(t)$  is a rotating field of amplitude  $H_1$  and of frequency  $\omega$ . The results of numerical simulation for the magnetization motion are presented in Fig. 1. The intuitive suppositions outlined above have been confirmed. To get clear picture of the magnetization motion it is convenient to exploit rotating coordinate system with X-axis coinciding with instant direction of RF field and Z-axis being perpendicular to the field rotation plane. In Fig. 1 we see the behaviour of magnetization evolution as frequency of rotating field increases. Note that the axis to which magnetization vector tends goes off the direction of rotating field and in limit of extremely large frequencies it nearly coincides with direction perpendicular to rotating field. It means that magnetization goes off the instant direction of the field. Let us define as a border frequency  $\omega_b = \gamma H_1$  at which the direction of magnetization makes an angle of  $45^\circ$  to the plane of field rotation. So for the field amplitude of  $H_1 = 1$  Oe this frequency is about 2.8 MHz. Hence for isotropic ferromagnetic substance there is a frequency depending upon the field amplitude, above which the field induced on nucleus differs considerably from the external field. The next step in analyzing the magnetization behavior is to account for interactions typical for magnetic substances. First of all these are the interactions describing the magnetic anisotropy fields. For example, the easy plane magnetic system behavior has its own peculiarities. Now magnetization is locked

in the easy plane. The adiabaticity regime has more distinct limits in this case. Such magnetization behavior at different frequencies and the same field amplitude is shown on Fig. 2 (Axial anisotropic field  $H_a = 1000$  Oe). Threshold frequency in this case means the change of the magnetization motion character. In low frequency case the magnetization catches the instant field direction (see Fig. 2 a,b) and in other case it gets torn from this direction (see Fig. 2 c,d). This means that easy plane systems have some advantages in controllable change of hyperfine magnetic fields. It should be noted that in easy plane case the threshold frequency shifts to the larger values as the constant of magnetic anisotropy (of easy plane type) increases. For example, for  $\text{FeBO}_3$  system ( $H_a$  is about  $6 \cdot 10^4$  Oe) this frequency is equal to about 700 MHz for the rotating field amplitude 10 Oe consuming that there is no direction in easy plane is singled out. This system is for a long time successfully used for fast switching of hyperfine field direction [15].

## Method of quasienergy spin states.

Rigorous interpretation of the effects appearing in Mossbauer spectra under the conditions of coherent dynamics of nuclear spins could be done on the basis of the quasienergy concept [16]. According to this concept [17-19] the state of a system with temporally periodic hamiltonian is characterized by quasienergy states. Obtaining these states for nucleus spin in arbitrary alternating hyperfine field is just a matter of calculation. These states being known the probabilities of spectral transitions between them are defined using the methods of calculation of such probabilities in case of stationary states. It is such an approach that will be used in this work for the exactly solvable model for rotating field. The quasienergy spin states may be defined in this case easily using, for example, the method described in [17,18]:

$$\Psi^\alpha = \exp(-iE_\alpha t) \sum_M d'_{\alpha M} |M\rangle \exp(i(M-\alpha)\omega t) \exp(-iM\varphi), \quad (1)$$

$$E_\alpha = -\omega M' + M' \left( (\omega_0 + \omega)^2 + \omega_1^2 \right)^{1/2} = -\omega M' + E_\alpha^0, \quad \alpha \equiv M',$$

$$\omega_0 = -\gamma H_0, \quad \omega_1 = -\gamma H_1, \quad d'_{\alpha M} = d'_{\alpha M}(\beta) - \text{rotation matrix}$$

$$\text{and } \beta = \arctg\left(\frac{\omega_1}{\omega_0 + \omega}\right), \quad \omega, \varphi - \text{frequency and phase of rotating field. } H_0$$

– constant hyperfine field perpendicular to field rotation plane.

Further we utilize the perturbation theory to analyze the spectroscopic transitions, but now instead of stationary basis states we use the quasienergetic states. The probability of transition of the nucleus due to interaction with the field of gamma-radiation,  $H_\gamma$ , from excited state  $\alpha$  to ground state  $\alpha'$  for a while  $\tau$ :

$$\begin{aligned} |a_{\alpha\alpha'}(\tau)|^2 &= \left| \int_0^\tau dt \sum_{Mm} d'^{le}_{\alpha M}{}^* \langle M | H_\gamma | m \rangle d'^{lg}_{\alpha' m} \right. \\ &\quad \left. \exp \left[ i \left( E_\alpha^0 - E_{\alpha'}^0 - \omega_\gamma \right) t - i(M-m)\omega t - i(M-m)\varphi \right] \right|^2, \end{aligned}$$

$$|a_{\alpha\alpha'}(\tau)|^2 = \left| \sum_{M-m} \sum_{M-m \neq \text{const}} d_{\alpha M}^{le} * d_{\alpha' m}^{lg} \langle M | H_\gamma | m \rangle \right| \quad (2)$$

$$\exp(-\lambda(M-m)\varphi) \Phi(E_{\alpha M} - E_{\alpha' m} - \omega_\gamma)^2$$

$$\Phi(E_{\alpha M} - E_{\alpha' m} - \omega_\gamma) = \frac{(\exp(\lambda(E_{\alpha M} - E_{\alpha' m} - \omega_\gamma)\tau) - 1)}{\lambda(E_{\alpha M} - E_{\alpha' m} - \omega_\gamma)}.$$

Here we introduced the notions:  $E_{\alpha M} = E_\alpha^0 - \omega' M$ ,  $E_{\alpha' m} = E_{\alpha'}^0 - \omega' m$ .

The given expression allows to obtain the probability of induced gamma-emission. If time-averaged registration of gamma-quanta is used this probability per unit of time is

$$P_{\alpha\alpha'}(k, \Sigma) = 2\pi \sum_{M-m} \left| \sum_{M-m \neq \text{const}} \langle I^e, M | H_\gamma(k, \Sigma) | I^g, m \rangle d_{\alpha M}^{le} * d_{\alpha' m}^{lg} \right|^2 \delta(E_\alpha^0 - E_{\alpha'}^0 - \omega_\gamma - (M-m)\omega) \quad (3)$$

$\Sigma$  indicates the polarization state of quantum inducing this transition.

The formula (3) defines the total number of spectral lines and their positions.

The more detailed information must be obtained taking into account the finite excited state lifetime and assuming that time filtration regime of emitted quanta registration is used.

Now

$$\Phi(E_{\alpha M} - E_{\alpha' m} - \omega_\gamma) = \frac{(\exp(\lambda(E_{\alpha M} - E_{\alpha' m} - \omega_\gamma)\tau - \frac{\gamma\tau}{2}) - 1)}{\lambda(E_{\alpha M} - E_{\alpha' m} - \omega_\gamma - \frac{\gamma\tau}{2})}.$$

The expression (2) we rewrite down singling out explicitly the terms of two types

$$\begin{aligned} |a_{\alpha\alpha'}(\tau)|^2 &= \sum_{M-m} \left| \sum_{M-m \neq \text{const}} d_{\alpha M}^{le} * d_{\alpha' m}^{lg} \langle M | H_\gamma | m \rangle \right|^2 F(\alpha M - \alpha' m) + \\ &+ \sum_{\Delta M \neq \Delta M'} \sum_{M-m \neq \Delta M} (...) \left( \sum_{M-m \neq \Delta M'} (...) \right)^* \Phi(E_\alpha^0 - E_{\alpha'}^0 - \Delta M \omega - \omega_\gamma) \Phi^*(E_\alpha^0 - E_{\alpha'}^0 - \Delta M' \omega - \omega_\gamma) \end{aligned} \quad (4)$$

The first sum does not depend on phase of the external alternating field. It describes the emission spectrum obtained by time-

averaged measurements. The second sum describes the time-dependent probability of transition. To make more clear the time filtration condition we write down the expression for probability of gamma-transition per unit of time:

$$P_{\alpha\alpha'}(k, \Sigma) = \int_0^{\infty} d\tau \frac{d|a_{\alpha\alpha'}(\tau)|^2}{d\tau}.$$

Actually, we measure the induced radiation at time  $t$  stipulated by a number of nuclear gamma-transitions in the time interval from  $-\infty$  to  $t$ .

Let us illustrate the behavior of the summands in (4) for  $\text{Fe}^{57}$  isotope. Accounting for magnetic-dipole transitions only we can easily demonstrate that in case of induced gamma-transitions the circularly-polarized radiation does not lead to temporal beats, while linearly-polarized radiation induces radiation with time-dependent amplitude. The behavior of induced radiation intensity in this latter case can be described by an expression:

$$P_{\alpha\alpha'} = P_{\alpha\alpha'}^0 + P_{\alpha\alpha'}(t),$$

$$P_{\alpha\alpha'}^0 \propto \sum_{\Delta M = \pm 1} \frac{1}{(E_{\alpha}^0 - E_{\alpha'}^0 - \Delta M\omega - \omega_{\gamma})^2 + \gamma^2/4},$$

$$P_{\alpha\alpha'}(t) \propto \frac{\sin 2\omega t}{\gamma} \left\{ \frac{-[(E_{\alpha}^0 - E_{\alpha'}^0)^2 - \omega^2 + \gamma^2/4]4\omega + 2\omega\gamma^2}{[(E_{\alpha}^0 - E_{\alpha'}^0)^2 - \omega^2 + \gamma^2/4]^2 + \omega^2\gamma^2} \right\}.$$

The behavior of  $P_{\alpha\alpha'}$  is shown on Fig. 3. for various values of  $\varphi = 2\omega t$ . Somewhat different result is obtained for the spontaneous gamma-quantum emission. The transition is then induced by zero oscillations of electromagnetic vacuum and we obtain the gamma-radiation as a linear combination of the photon states of different polarizations and energies. Such composite photon state can be imagined as a generalization of known photon states being linear combinations of photon states with different polarizations but of the same energies.

### The common discussion of the problem.

RF effects in gamma-optics are today closely related to the possibility of achieving inversionless gamma-radiation amplification regime. The analysis of such possibility was carried out recently [20] on the base of density matrix formalism. The necessary condition of inversionless amplification is to achieve the destructive interference which leads to nonreciprocity of gamma transitions. In quasienergy states representation the radiation transition parameters acquire more distinct structure. First of all, the coherence induced by RF field and necessary for destructive interference is now included in quasienergy states themselves. It is well known [19] that quasienergy states form a complete set of states and may serve as a basis for density matrix representation. The density matrix

equations look simpler in this case, though they may contain the terms describing dissipative processes too. The systems of density matrix equations written in stationary states representation and in quasienergy states one may describe the same physical situation, but there is at least one essential difference. This concerns the difference between quasienergy states energies and stationary states ones. Let us, for example, consider a three-level system (Fig. 4). The  $\varphi_2$  and  $\varphi_3$  states (with energies a and b) are placed close to each other and are connected to the ground state  $\varphi_1$  by strong RF field of frequency  $\omega$ .

The relations  $\omega = \frac{a+b}{2}$  and  $\varepsilon_0 = \frac{b-a}{2}$  being true the following quasienergy states are obtained ( $\varepsilon = E_\alpha$ ):

$$\psi_{\varepsilon=0} = N_0 \left( \frac{\varepsilon_0}{d} |\varphi_1\rangle + \exp(-i\omega t) (|\varphi_2\rangle + |\varphi_3\rangle) \right),$$

$$\psi_{\varepsilon=-D} = N_+ \exp(iDt) \left( \frac{-2d}{D+\varepsilon_0} |\varphi_1\rangle + \exp(-i\omega t) \left( |\varphi_2\rangle - \frac{D-\varepsilon_0}{D+\varepsilon_0} |\varphi_3\rangle \right) \right),$$

$$\psi_{\varepsilon=D} = N_- \exp(-iDt) \left( \frac{2d}{D-\varepsilon_0} |\varphi_1\rangle + \exp(-i\omega t) \left( |\varphi_2\rangle - \frac{D+\varepsilon_0}{D-\varepsilon_0} |\varphi_3\rangle \right) \right).$$

Here  $N_0, N_+, N_-$  are the normalizing coefficients,  $D = (2d^2 + \varepsilon_0^2)^{\frac{1}{2}}$ ,  $d$  is a matrix element of dipole interaction between the system and the RF field. The peculiarity of the given system of wave functions is that these functions are represented by coherent superposition of initial states with states modified by the well-known strong field effect. This circumstance may introduce essential corrections to analysis of inversionless amplification regime possibility.

The possibilities and requirements of controlled magnetization dynamics of magnetic system in external RF fields were studied in this work. The study of temporal and spectral changes in gamma-transition spectra was another subject of our investigation. These investigations supplement recently published results [7]. The study of pulse RF field influence on system magnetization and, finally, on nuclear spin is a next step in the investigations in this field.

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Signature

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## Captions to figures.

Fig. 1. The behavior of the magnetization  $M(t)$  ( $|M(t)|=1$ ,  $\alpha=l=0.1$ ,  $H_1=H^*H_0=10$  Oe,  $H_a=2K|M(t)|=H_a^*H_0=0$ ,  $H_0=10$  Oe) in the rotating coordinate system.  $M_x$ ,  $M_y$ ,  $M_z$  are projected along x, y, z axes. Initial conditions:  $M(0)=\left(\frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0\right)$ . (a,b) -  $\omega=\gamma H_0\omega'=2\pi*10$  sec $^{-1}$ ; (c,d) -  $\omega=\gamma H_0\omega'=2\pi*100$  sec $^{-1}$ .

Fig. 2. The behavior of the magnetization  $M(t)$  ( $|M(t)|=1$ ,  $\alpha=l=0.1$ ,  $H_1=H^*H_0=10$  Oe,  $H_a=2K|M(t)|=H_a^*H_0=1000$  Oe,  $H_0=10$  Oe) in the rotating coordinate system.  $M_x$ ,  $M_y$ ,  $M_z$  are projected along x, y, z axes. Initial conditions:  $M(0)=\left(\frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0\right)$ . (a,b) -  $\omega=\gamma H_0\omega'=2\pi*10$  sec $^{-1}$ ; (c,d) -  $\omega=\gamma H_0\omega'=2\pi*600$  sec $^{-1}$ .

Fig. 3. The emission probability  $I=P_{\alpha\alpha'}$  induced by linearly polarized probe gamma-radiation in RF excited spin system as function of time:

$\varphi=2\omega t=0$  (a),  $\varphi=\pi/2$  (b),  $\varphi=\pi$  (c),  $\varphi=3\pi/2$  (d).

Fig. 4. The energy structure of quasienergy states of three-level system in resonant field.

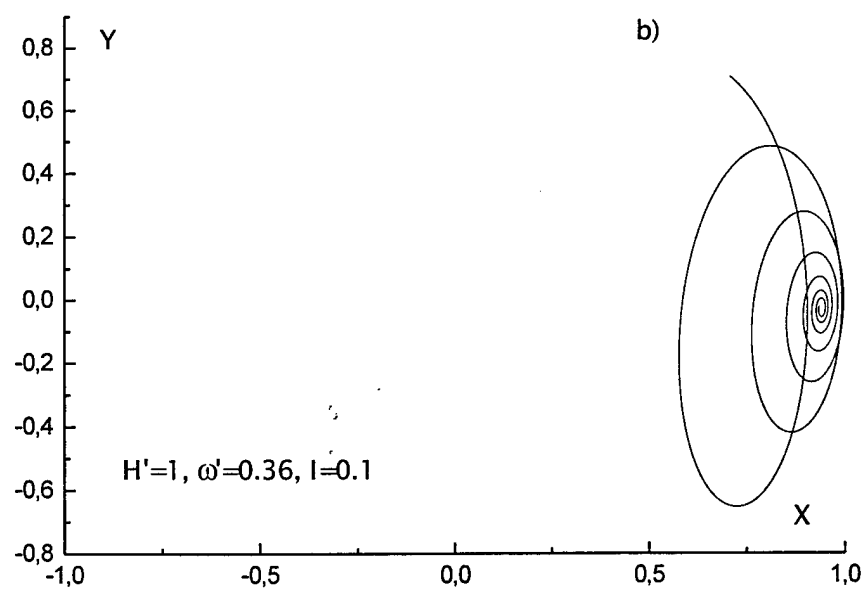
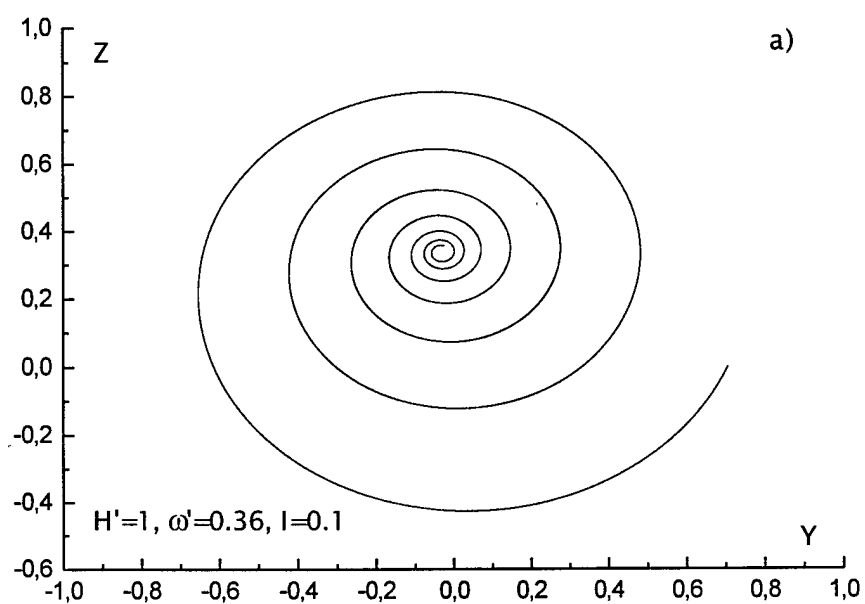


Fig. 1 a,b

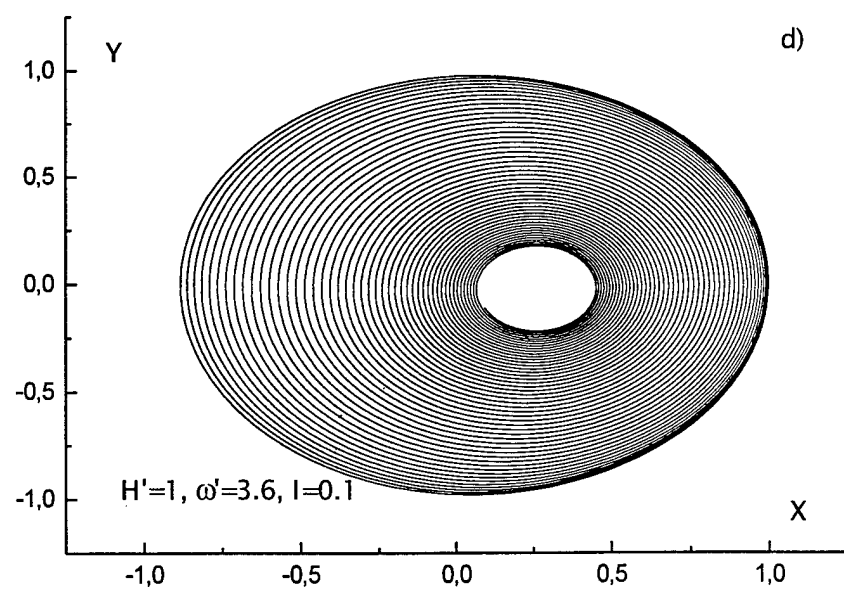
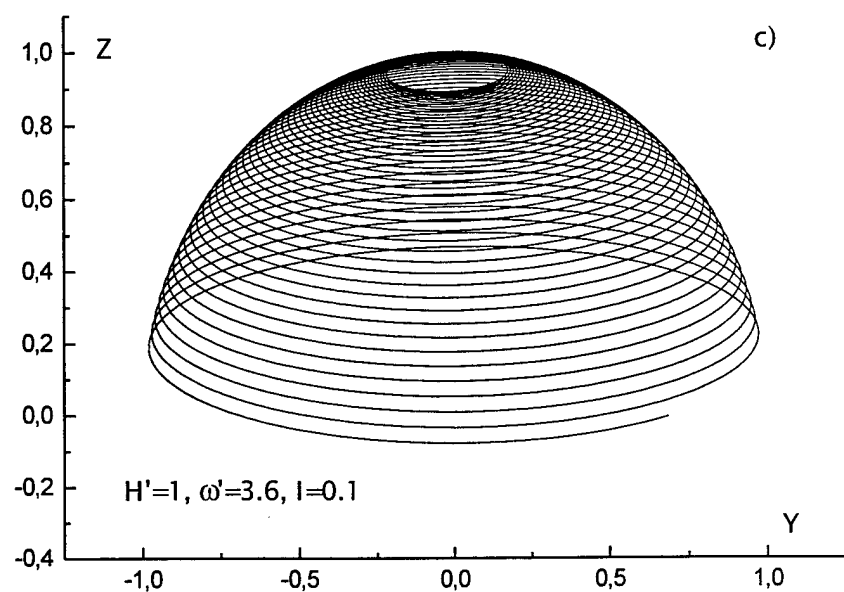


Fig. 1 c,d

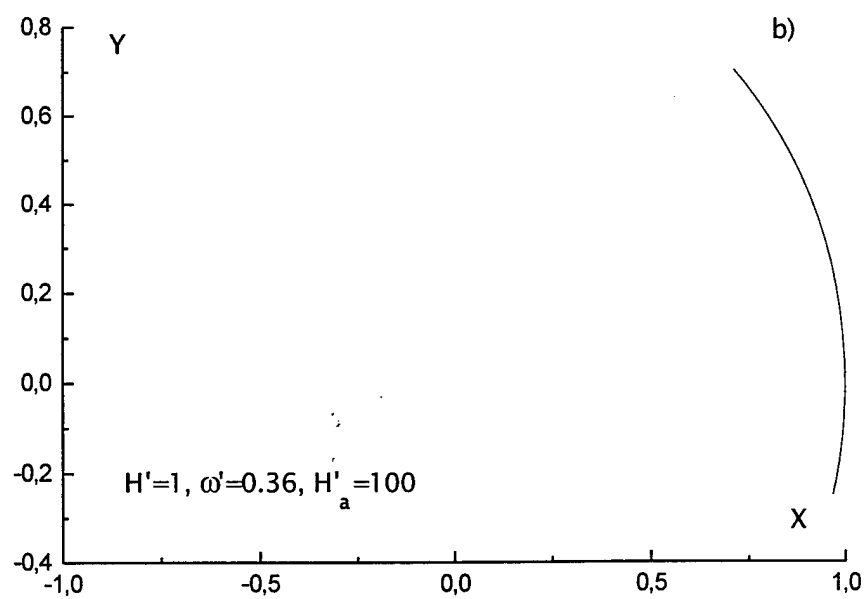
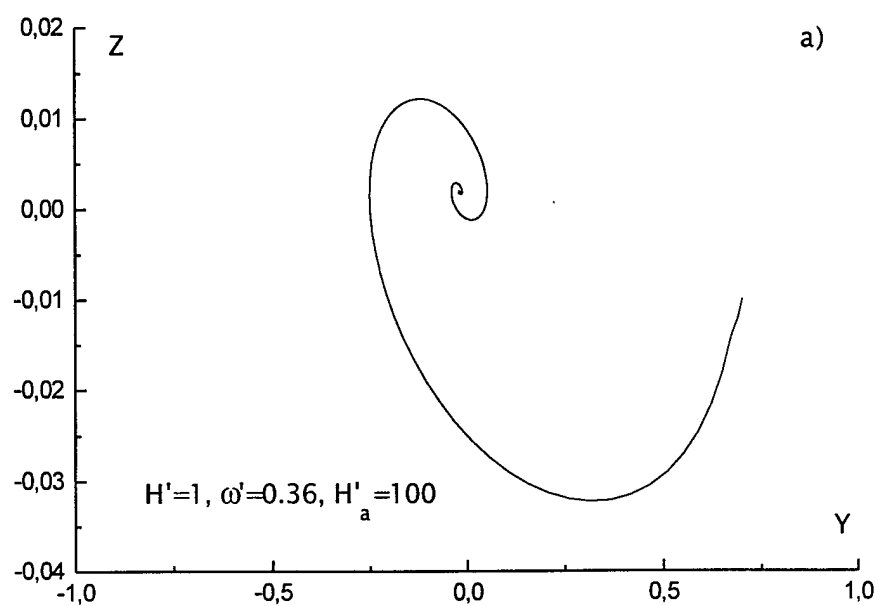


Fig. 2 a,b.

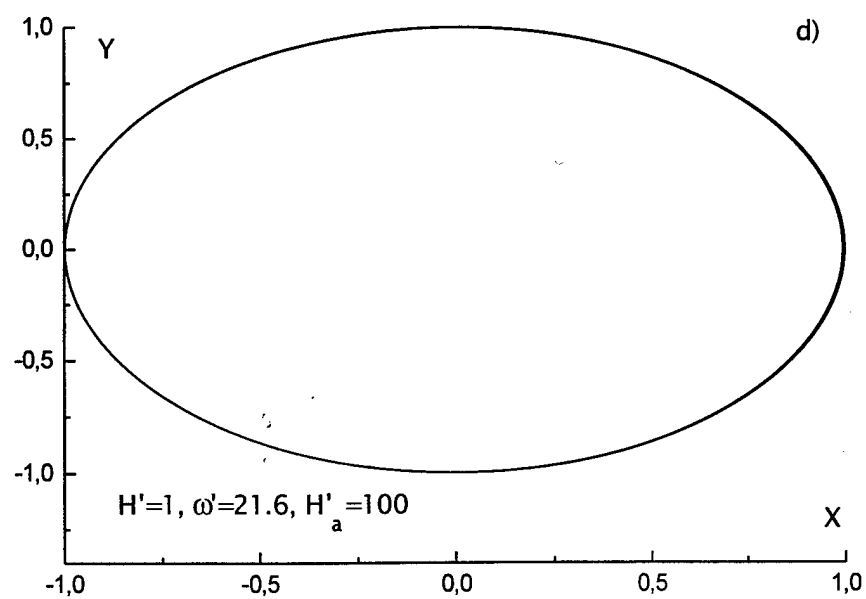
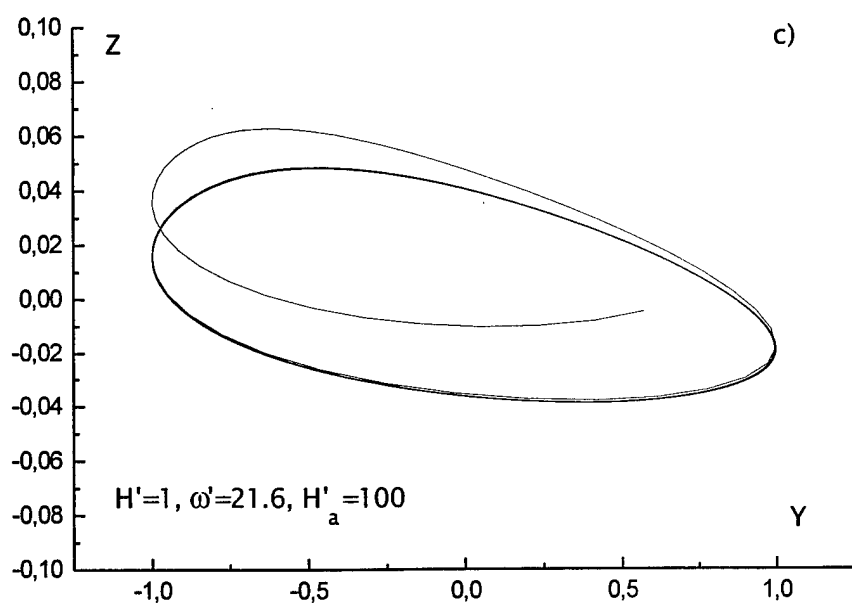


Fig. 2 c,d.

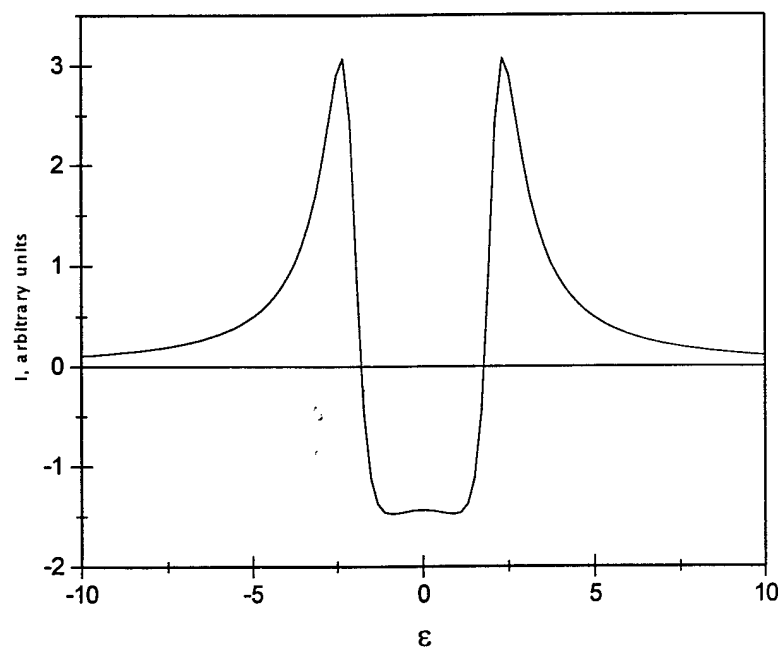
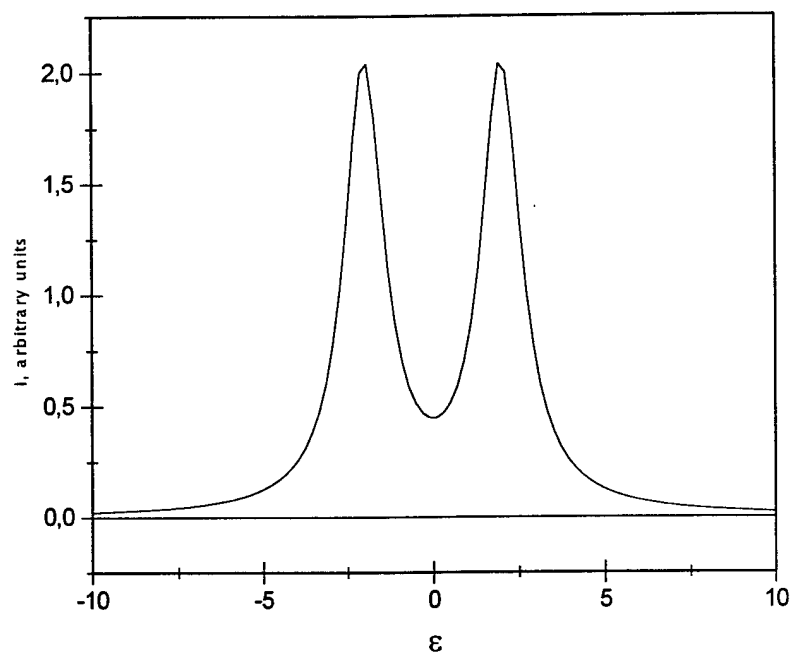
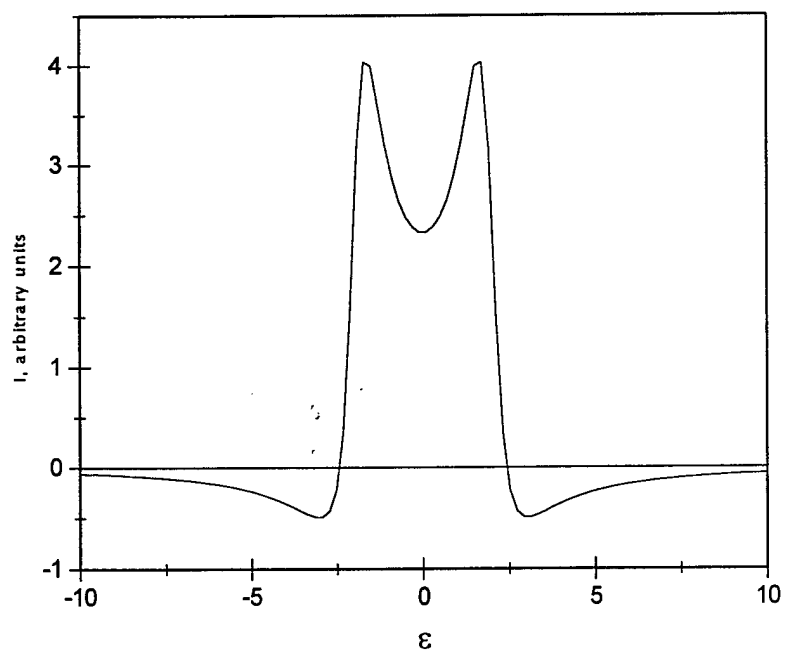
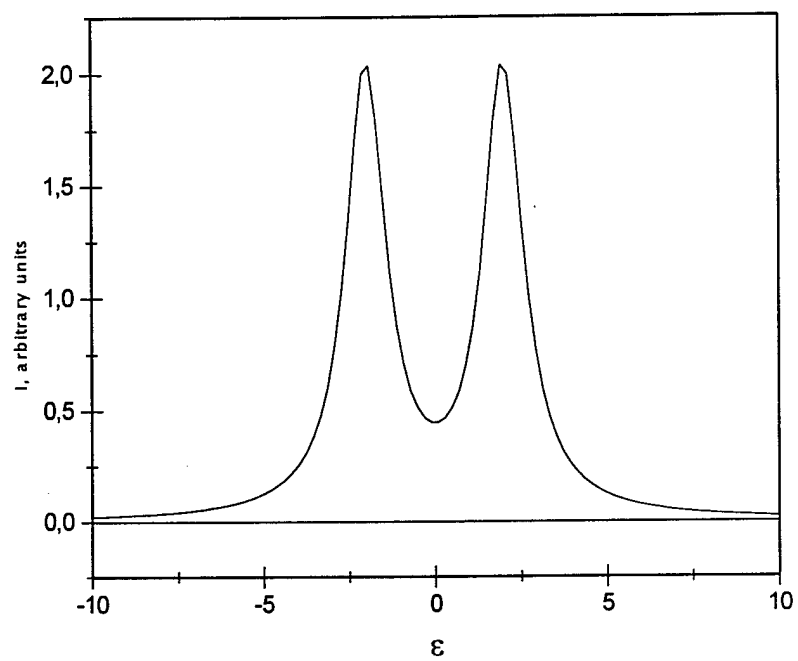


Fig. 3.



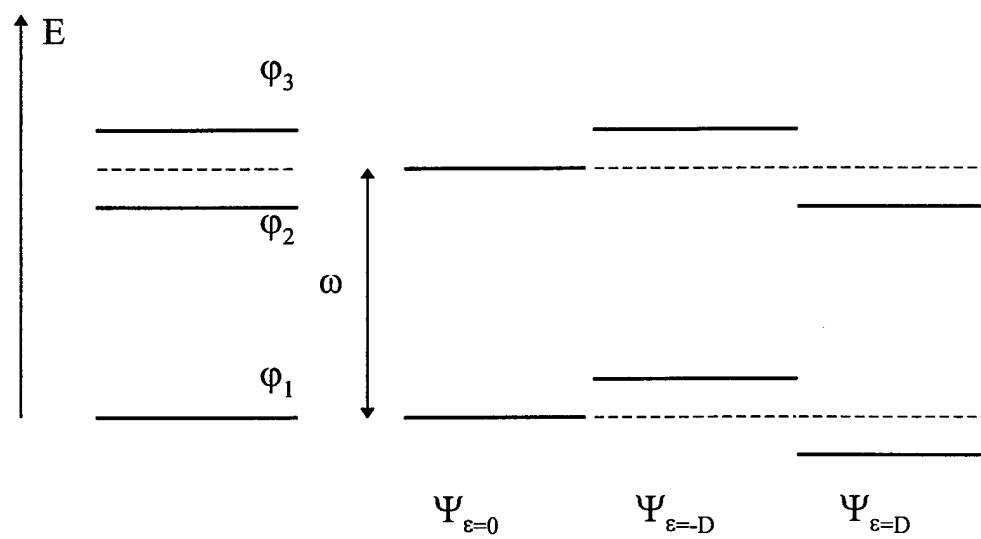


Fig.4